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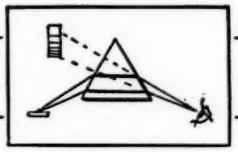

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An Experimental Study of the Triboluminescence of Certain Natural Crystals and Synthetically Prepared Materials

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(Received June 7, 1937)

TRIBOLUMINESCENCE, sometimes called "trennungsleuchten," has been the subject of a considerable amount of research since the summaries of work done along this line were published by Imhof,[1] and by Lenard,[2] Schmidt and Tomaschek. As is well known, triboluminescence[3] is often associated with other manifestations of luminescence but comparatively few experimental observations have been made to show exactly how these various phenomena, observed in the same material, are related to each other. Waggoner,[4] in some visual observations and Nelson,[5] photographically, have studied the tl spectra of a number of substances and have found the same broad bands characteristic of the fluorescence and phosphorescence of these same materials. Because of the dimness of TL and because of its short duration spectroscopic observations are difficult and not many have been made. Przibram[6] and his associates have observed that the intensity of the TL emitted by such crystals as rocksalt and fluorite, when they are ground in a mortar or crushed in a vise, is greatly increased by exposure to radium. The observations here described were made upon the TL of fluorite and other substances including synthetically prepared materials before and after exposure to radium and special attention is given to the relation of TL to thermoluminescence.

The work[7] was done at the Institute for Radium Research in Vienna, with help and many suggestions from Professor Karl Przibram during the entire course of the experiments. The author wishes to express her sincere appreciation of the cordial hospitality and cooperation shown her by Professor Meyer and Professor Przibram and of the generous way in which the facilities of the Radium Institute were placed at her disposal.

FLUORITE

From a large collection of fluorites, those listed in Table I were selected for study. All of these were tl in the natural condition and the TL was greatly intensified by exposure, for periods varying from 18 to 24 hours, to radium, standard preparation No. 5 (610 mg), at a distance of a few cm.

[1] A. Imhof, Physik. Zeits. **18**, 78 (1917).
[2] *Handbuch der Experimental Physik*, Vol. 23, 976.
[3] In this paper the abbreviations suggested by Imhof are used as follows: Triboluminescence—TL, Triboluminescent—tl.
[4] C. W. Waggoner, Phys. Rev. **7**, 402 (1916).
[5] D. M. Nelson, J. O. S. A. **12**, 207 (1926).
[6] K. Przibram, Mitt. d. Inst. f. Radiumforschung, No. 196, Wien. Ber. 11a, **136**, 43 (1927). K. Przibram and Kara Michailowa, Mitt. d. Inst. f. Radiumforschung, No. 149, Wien. Ber. 11a, **131**, 511 (1922). F. Urbach, Mitt. d. Inst. f. Radiumforschung, No. 261, Wien. Ber. 11a, **139**, 353 (1930).
[7] Frances G. Wick, Mitt. d. Inst. f. Radiumforschung, No. 389, Wien. Ber. 11a, **145**, 689 (1936).

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The TL of all fluorites, both before and after exposure to radium, was green in color and varied in intensity with the specimens and with the time of exposure to radium and the period elapsing between the end of the exposure and the observation. The TL could be seen when the crystals were ground in a mortar, crushed in a vise, scratched with a file or ground on a stone grindstone.

Spectrum Observations

Visual observations were made of the tl spectra of all the fluorites listed in Table I after exposure to radium. The spectra of some of the specimens were also observable without exposure to radium and were found to be the same as in the case of the same materials after exposure but less intense. For spectrum observations the crystals were ground in a mortar or crushed in a vise in front of the slit of a direct vision luminescence spectroscope made by Schmidt and Haench. The dispersion of the instrument is small and the observations do not have a high degree of precision. The spectra observed in specimens 1 through 11, Table I, were not continuous but consisted of narrow, well-defined bands such as are characteristic of the fluorescence, phosphorescence and thermoluminescence spectra of the fluorites and are due to rare earth impurities.[8–12]

Table I. *Fluorite.*

Number	Source	Natural Color
1	Axel Alpe, Pinzgau, near Salzburg	White
2	Montrose, California	Green
3	New Mexico, near Deming	Green
4	New South Wales	Green
5	Obira, Japan	Pale green
6	Weardale, Durham, England	Blue-green
7	Weardale, Durham, England	Purple
8	Westmoreland, New Hampshire	Green
9	Unknown, A	Green
10	Unknown, B	Pale green
11	Trumbull County, Conn. Chlorophane	Reddish brown
12	Franklin, New Jersey	Reddish brown

[8] Urbain, Ann. de Chemie et de Phys. **18**, 222 (1909).
[9] Haberlandt, Karlik, Przibram, Mitt. d. Inst. f. Radiumforschung, No. 336, Wien. Ber. 11a, **143**, 151 (1934).
[10] F. G. Wick, Phys. Rev. **24**, 272 (1924).
[11] H. Steinmetz, Naturwiss. **24**, 172 (1936).
[12] H. Haberlandt, Wien. Ber. 11a, **144**, 663 (1936).

The tl spectra of all the fluorites showed strong bands or groups of bands at wave-lengths 5400 to 5700A and 4500 to 5000A and, in many cases, a weaker band between 4300 and 4400A. Comparison with the work of Urbain[8] shows that the band 5400 to 5500A is the strongest band of Tb, and 4300 to 4400 a weaker band of Tb. Between 4500 and 5000A are strong bands of Dy and weaker bands of Tb. The relative intensities of the tl bands vary greatly with the different specimens. Specimens 8 and 9, Table I, show the Tb band, 5400 to 5500A, most strongly and 7 and 8 show the Dy band, 4800 to 5000, very strongly. A few of the fluorites, especially those which, in the natural condition show strong green thermoluminescence, such as specimens 11 and 12, Table I, show, in addition to the narrow rare earth bands, a continuous green band extending from 4500 to 6000A after exposure to radium. This band is characteristic of Mn. Specimen 12, which contains a high percentage of Mn, shows only this broad green band in the tl spectrum.

Relation of Triboluminescence to Phosphorescence and Thermoluminescence

The fluorites observed were all phosphorescent after exposure to radium. In most cases the phosphorescence was too dim for spectroscopic observation but in the case of specimen 8, Table I, the phosphorescence spectrum was observed to be the same as that of the TL. The color of the TL in all the specimens was the same as that of the phosphorescence.

Thermoluminescence, which is very strong in fluorite after exposure to radium, was observed by grinding the material coarsely then heating it slowly on a copper plate over a Bunsen burner. In general, there is an intense green emission which comes up quickly at a low temperature and lasts a very short time; this is followed at an intermediate temperature, by an emission of a different color, in many cases purplish, which is more persistent. At still higher temperatures there is another distinct color change, very often a return to the green color emitted at the lowest temperature upon first heating. The high temperature emission is much less intense than the low.

Spectroscopic observations show, in the thermoluminescence which appears first after the heating is begun, the same rare earth bands which are seen in the TL. The spectrum of the purplish thermoluminescence which follows at intermediate temperatures shows bands characteristic of other rare earths especially Sm and Eu and also the yellow and red bands of Dy and Tb. At still higher temperatures there is another color change in the thermoluminescence, the spectrum, in many cases, showing the bands seen at the lowest temperature. In a number of specimens a strong continuous band due to Mn appears after the lowest temperature emission and before the spectrum characteristic of intermediate temperatures. Specimen 12, Table I, differs from the others in that the spectrum of the natural material as well as that after exposure to radium, at lowest temperatures, shows only the broad continuous band of Mn. After exposure to radium, at intermediate temperatures, the red band of Sm, 6040A, and the yellow band of Dy, 5765A, can be seen. Other rare earth bands are probably present but obscured by the strong Mn band.

Because the tl spectrum is identical with that of the lowest temperature thermoluminescence, the question arises as to whether or not the specimens in which Tl was observed may have been sufficiently heated in the process of grinding or crushing for the emission of ordinary thermoluminescence. Because direct temperature observations at the instant at which a crushed crystal gives off light are impossible, some observations were made, at the suggestion of Professor Przibram, from which the rise of temperature due to the work done in crushing the crystal could be computed, assuming that the crystal was uniformly heated. A weighed piece of crystal No. 9, Table I, was crushed in a vise by attaching a known weight to the handle of the vise and allowing this weight to drop through a measured distance sufficient to crush the crystal causing the emission of TL. The heat equivalent of the work done was computed and, taking into consideration the mass and specific heat of the crystal, it was found that the heat developed would be sufficient to raise the temperature of the crushed crystal 5°C, assuming that the heat all went into the crystal and heated it uniformly. Observations of this same crystal showed that the first emission of thermoluminescence came at 160°C which is 140°C above the temperature of the room. It appears evident that TL is not a true emission of thermoluminescence.

Effect of Heating Crystals in Blast Before Crushing

It has been previously observed, as stated by Imhof,[1] that fluorites and other crystals show TL after they have been heated to a temperature sufficient to exhaust the thermoluminescence. In this work the crystals were heated still higher than the temperatures recorded previously. In some cases they were maintained at red heat for half an hour but generally the crystals were heated in a quartz crucible in a blast for a shorter time. After the crystals were cooled they still showed TL which was less intense than that shown by the natural crystals or by those exposed to radium. The heated crystals showed TL which was strong enough for spectroscopic observation when they were crushed in a vise. The same material crushed in a mortar appeared considerably less bright.

Specimens 4 and 8, Table I, after heat treatment, show the same two bands in the TL which were observed in the unheated specimens. Specimen 7, which contains a relatively high percentage of Eu[10] and less Tb than specimens 4 and 8, shows a very strong band from 4200 to 5500A which is characteristic of Eu, superimposed upon which are bands of Tb and Dy. It has been shown by Przibram[13] and his associates that, after prolonged heating, the blue fluorescence due to Eu cannot be excited by ultraviolet light but that exposure to radium regenerates fluorite crystals containing Eu so that this fluorescence can again be excited. Such crystals are said to show "radiophoroluminescence."

It is evident that the fluorites emit TL which is not dependent upon the excitation necessary for thermoluminescence. This TL varies in different specimens depending upon the impurities present.

[13] Haberlandt and Przibram, Mitt. d. Inst. f. Radiumforschung 313, 11a, **142**, 235 (1933).

Effect of Grinding Crystals Under Liquids

That the TL of some substances is due, in part at least, to an electric discharge in air is known.[2] Longchambon[14] photographed the tl spectra of a number of substances one of which was fluorite and found many bands characteristic of nitrogen. It was suggested by Przibram that any part of the TL of fluorite which was due to an electric discharge in air would be modified if a medium of a different character and conductivity surrounded the crystal. In order to test this, fluorite crystals were ground under xylol and under water. The TL of the natural crystals and of those exposed to radium was somewhat, though not greatly, dimmer than in air indicating that a part but not a high percentage of this TL might be due to an electric discharge in air. The spectrum of specimen 7, Table I, looked somewhat sharper than it did in air. The crystals heated in the blast before grinding had the intensity of TL much reduced under liquids indicating that a considerable part of the light but not all of it might be due to an electric discharge in air.

Effect of Low Temperature

The intensity of TL has been observed by Imhof[1] to vary with temperature, usually diminishing as the temperature of the specimen is raised. His observations upon a specimen of natural fluorite indicate no change in the intensity of TL when the temperature is lowered to $-80°C$. Low temperature observations were made upon a few of the crystals listed in Table I by suspending the specimens in liquid air then crushing them in a vise allowing the minimum of time for temperature change. Crystals exposed to radium showed stronger TL at room temperature than at low temperature. Crystals previously heated in the blast showed no change in the TL. It seems probable that the centers which are affected by change in temperature are completely deactivated by the high temperature of the blast and that the electric discharge in air is not modified by a change in temperature of the crystal. Wick and Carter[15] observed that a change in temperature during exposure of fluorite to high voltage cathode rays makes no difference in the thermoluminescence resulting from such exposure.

Tribothermoluminescence

A mechanical method of exciting material to thermoluminescence was observed by Nyswander and Cohn[16] for a number of substances including glass and colorless fluorite. The materials were heated to redness then ground in a mortar. The powdered material showed a dim thermoluminescence when heated to a temperature of about 350°C which was called "tribothermoluminescence." All of the fluorites listed in Table I showed this effect after the heated material was ground in a mortar then heated in an electric furnace or upon a copper plate. This tribothermoluminescence appeared at a high temperature and persisted, in many cases, until red heat had been reached. This makes it appear possible that triboluminescence may be due to an activation, by mechanical means, of the same centers which emit the highest temperature thermoluminescence. The intensity of the tribothermoluminescence was not high enough for spectroscopic observations. That tribothermoluminescence was not due to exposure of the heated crystals to light was proved by heating some crystals of specimen 9, Table I, exposing part of them to light for four days, keeping the other part in the dark. No difference could be observed in the emission.

Size of Grain

It is well known that size of grain makes a difference in the intensity of TL. The minimum size, under which no TL can be observed, varies with different materials. Imhof[1] gives a table for different materials in which the minimum for fluorite is given as 0.7 to 1.2 mm. Observations upon the fluorites in Table I show that TL may be observed in much finer grained material. A number of these fluorites were heated red hot in a blast, then pulverized by pounding the material on an iron plate with an iron hammer, heated again, then sifted through gauze of varying size of mesh. Specimen 7 showed TL when the par-

[14] Longchambon, Comptes rendus **174**, 1633 (1922). Comptes rendus **176**, 691 (1923).

[15] Wick and Carter, J. O. S. A. **18**, 383 (1929).

[16] Nyswander and Cohn, Phys. Rev. **36**, 1257 (1930).

ticles were sifted through a gauze the openings of which were 0.05 mm; specimens 4 and 8 when sifted through openings of size 0.12 mm. Since the fluorites show considerable difference in their TL an attempt was made to find one which, in the natural state, without being subjected to high temperature, would show a minimum size of grain comparable with that given by Imhof[1] for a specimen described as "white." Clear colorless fluorites from Sarnthal and from Franklin, N. J. were sifted between two gauzes of mesh sizes 0.16 and 0.12 mm and both of them still showed TL. The powdered specimens were then heated to redness in the blast and showed stronger TL after heating than before.

It is well known that the intensity of luminescence of materials, in general, is modified by subjection to high temperature and that fluorite, heated before exposure to cathode rays,[10] shows brighter cathodoluminescence than the same fluorite unheated. Since TL exhibits many of the characteristics of other forms of luminescence it is not surprising that previous heat treatment should change the intensity of TL shown by specimens in which the method of excitation is mechanical breaking.

The effect of exposure to radium appears to depend upon size of grain. In order to test this, specimens 3, 4, 7, and 8, Table I, were sifted to seven different sizes between sieves varying in mesh from 0.73 mm to 0.05 mm. After an exposure to radium of 17 hours it was found that the TL, phosphorescence, and thermoluminescence of the coarsest grained material was strongest and of the finest, weakest. The period of phosphorescence diminished with the size of grain. More prolonged exposure to radium increased the difference in period between the grains of different size. After an exposure of 154 hours the phosphorescence of the coarse grained material was weaker than after a shorter exposure and the fine grained material showed practically no phosphorescence. The TL and the low temperature thermoluminescence were also much weaker in the fine grained material. The high temperature thermoluminescence showed less difference. Professor Przibram suggests that the difference in the effect of exposure to radium upon fine and coarse grained material may be due to the effect of pressure used in pulverizing[6] or possibly to the fact that the larger grains become more highly colored than the smaller and absorb more of the emitted light. The larger crystals were colored violet; the smaller ones looked opaque and almost white because of the powdered condition.

Work Upon Synthetic Materials Containing CaF_2 and the Rare Earths

Synthetically prepared materials of CaF_2 and small amounts of the different rare earths have been found by Urbain,[8] Przibram,[9] and others to show the same luminescence bands under excitation by cathode rays and ultraviolet light and in thermoluminescence as those seen in the natural specimens of fluorite. A number of such preparations, kindly made by Dr. Rona, show that the TL also may be duplicated in synthetically prepared materials. A list of the preparations examined is given in Table II.

All of the preparations listed in Table II show TL strong enough for spectroscopic observations after exposure to radium but the preparations of CaF_2 plus Tb were stronger than the others. Preparation 5 (more Tb) was much stronger than preparation 4 (less Tb). It is also true that preparation 7 (more Mn) showed stronger TL than preparation 6 (less Mn). Preparations 1, 2, and 3, containing CaF_2 with small percentages of different rare earths, showed TL strong enough for spectroscopic observations but the thermoluminescence was too dim to permit such observations. A comparison of preparation 5 (1 percent Tb) with preparation 7 (1 percent Mn) showed that the thermoluminescence of the former came up at a lower temperature than that of the latter. All of the preparations in Table II which contained rare earths showed TL after heating without exposure to radium and also tribothermoluminescence. The preparations which contained Mn but no rare earths showed neither of these. Specimens 10 and 11, Table II (CaF_2 +0.5 percent Tb+0.5 percent Mn), appear to behave, in every way, like the natural fluorites, especially such a fluorite as specimen 11, Table I, which definitely shows the presence of Tb and Mn in its thermoluminescence spectrum. In both this fluorite and the synthetic material the low temperature thermoluminescence shows only the rare earth bands; at intermediate temperatures

TABLE II. *Synthetic preparations made of CaF_2 and rare earths.*

	PREPARATION	PREPARATION HEATED RED IN BLAST, THEN EXPOSED TO RADIUM	
		TL	THERMOLUMINESCENCE
1	CaF_2+0.01% Dy	Green, Dy bands 4700–4800A 5500–5800A	Green, dim. Too dim for spectroscopic observations
2	CaF_2+0.01% Sm	Orange, Sm band strong 5500–6000A	Orange, dim. Too dim for spectroscopic observations
3	$CaSO_4$+0.01% Sm	Very strong orange, Sm bands 5500–5800A 6000A	Strong, orange. Three very sharp bands of Sm 5600, 6000, 6400A
4	CaF_2+0.017% Tb	Green, strong. Strongest Tb band 5400–5500A	Green, dim
5	CaF_2+1% Tb	Green, very strong. Tb bands 5400–5500 very bright 4800–5000 medium 4400–4500 dim	Green, very strong. Tb bands
6	CaF_2+0.017 Mn	Mn band, green 4700–5500A	Mn band, strong green
7	CaF_2+1% Mn	Mn band, green stronger than 6	Mn band, green, stronger than 6, came up at a lower temperature
8	$CaSO_4$+0.4% Mn	Mn band, green, strong	Mn band, green, strong
9	$CaSO_4$+0.8% Mn	Mn band, green, less strong than 8	Mn band, green, less strong than 8
10	CaF_2+0.5% Tb +0.5% Mn	After 24 hour exposure to radium, strong green, rare earth bands only 4800–5000A 5400–5500A	After 24 hour exposure to radium, strong green, only Tb bands
11	CaF_2+0.5% Tb +0.5% Mn	After 225 hour exposure to radium 5400–5500A, strong 4800–5000A, weaker Mn bands, dim	After 225 hour exposure to radium. Tb bands, then Mn band, then again Tb bands

the Mn band appears very strongly and at a higher temperature the Mn band disappears and the Tb bands are again prominent. The Tb spectrum in specimen 10, and in specimen 5, Table II, shows two distinct maxima at different temperatures. The Tb bands can be seen to come up very quickly in specimen 5 as the material is warmed on a copper plate, then these bands definitely weaken and, after the temperature is considerably higher, the Tb bands again brighten more gradually and are more persistent but not so bright as in the lower temperature maximum. Two distinct thermoluminescence maxima have been observed in synthetic materials such as $CdSO_4$+Mn and $ZnSO_4$+Mn after exposure to x-rays.[17] It is interesting to note also that candoluminescence of aluminum oxide containing Tb, as observed by Nichols and Wick,[18] shows two distinct maxima under a temperature of 400°C.

Exposure to radium or x-rays appears to activate two kinds of centers as described by Lenard, one easily disturbed, unstable, and manifesting itself by phosphorescence and low temperature thermoluminescence, the other, much more stable, manifesting itself as high temperature thermoluminescence.

DOMINANCE OF CERTAIN RARE EARTHS IN TL SPECTRA

It is well known[8] that in luminescent specimens containing different impurities some of these are

[17] Wick and Slattery, J. O. S. A. **16**, 398 (1928).

[18] Nichols and Wick, J. O. S. A. **22**, 357 (1932).

known to be dominant and to suppress the effects of others. Nichols[19] observed that, under cathode rays, CaO+Mn gives the Mn band.

TABLE III. *Calcite.*

NUMBER	SOURCE	COLOR
1	Joplin, Missouri	Yellowish
2	Rabenstein, Saxony	Slightly yellowish
3	Habachthal, Salzburg	Very slightly yellowish
4	Iceland	Clear, colorless

When any one of five rare earths is added (Th, Eu, Dy, Tb or Sm) the continuous spectrum of Mn disappears almost completely leaving only the line-like bands of the rare earth. He found that in solid solutions containing Tb and Dy, if Tb predominates, the Dy bands disappear; if Dy predominates both sets of bands appear.

The work here done upon synthetic materials shows that, in a specimen of CaF_2 containing equal percentages of Mn and Tb, specimen 10, Table II, the Tb dominates in the tl spectrum and in the low temperature thermoluminescence spectrum. In the spectra of the fluorites which are known to contain various other impurities the rare earths dominate in the tl spectra of every specimen observed except specimen 12, Table I, which contains a relatively large percentage of Mn. That rare earths also are present in this specimen was observed in the high temperature thermoluminescence which showed the red and yellow bands of Sm and Dy. The bands of shorter wave-length were obscured by the strong Mn band.

It was observed that the rare earths which dominate in the tl spectra are Tb and Dy. These are the dominant ones also in low temperature thermoluminescence and in phosphorescence. Some of the specimens examined were known to contain other rare earths in higher proportions. For example, blue-green fluorite from Weardale, specimen 6, Table I, shows most strongly, under cathode rays, the lines of Sm[10] and Gd and less strongly the lines of Tb, Dy, and Eu. In the purple fluorite from Weardale, specimen 7, Table I, Eu[10] predominates in the cathode ray spectrum. In TL only the bands of the element which predominates may be seen. In thermoluminescence the bands of the other rare earth elements which the specimen contains in lower concentration may also be seen at different temperatures. The cathodoluminescence spectrum of the fluorites is rich in bands because many bands which appear at various temperatures in thermoluminescence are observable at the same time under cathode-ray bombardment.

[19] Nichols and Howes, Carnegie Publication No. 384, p. 172 (1928).

TABLE IV. *Natural substances which show triboluminescence like that of fluorite.*

		NATURAL SUBSTANCE		EXPOSED TO RADIUM		HEATED IN BLAST	
	MATERIAL, SOURCE	TL	THERMO-LUMI-NESCENCE	TL	THERMOLUMINESCENCE	TL	THERMO-LUMI-NESCENCE
1	Apatit from Ep-prechtstein	Green, strong	Very weak	Green, very strong	Green, strong. Band spectrum 5000–6000A	Medium	Dim
2	Chalcedony amorphous	Medium	None	Strong, green, then red at high pressures	Very dim green	Strong	Dim
3	Perlsinter from a hot spring in Joskana	Red, good	None	Good, green, then red at high pressures	Very, very dim. Green at high temperatures	Strong	Dim
4	Rocksalt	Dim	Dim	Good	Good	Dim	Medium
5	Feldspar, Haddon Neck, Conn.	Green, dim	Green, very dim	Green, very strong	Green, strong at high temperatures	Green, strong	Good
6	Quartz, clear crystal	Very strong	None	Strong, greenish	Dim, blue-green, high temperature	Strong	None

Calcite

Four specimens of calcite from different localities as indicated in Table III showed TL which, in general, had the same characteristics as that of the fluorites.

They showed TL in the natural state which was intensified by exposure to radium, with the exception of specimen 4, Table III. They show Tl after they are heated in the blast and, with the exception of specimen 1, show tribothermoluminescence.

They differ from fluorite in that the color of the TL is green and that of the low temperature thermoluminescence following exposure to radium is orange. At higher temperatures the thermoluminescence is greenish-yellow, the intensity of the green component in the spectrum varying with the different specimens.

The calcites differ more among themselves than the fluorites do. This variation may be explained by the fact that the impurities in calcite found in different localities are not the same. W. P. Headden[20, 21] has found that different calcites contain many different impurities including the rare earths. The difference in the color of the TL from that of the low temperature thermoluminescence may be explained upon the basis of these impurities many of which are active in thermoluminescence and only one or a limited number of which, possibly the rare earths, are dominant in TL. The relative concentrations of the impurities may be such as to account for the differences[22] in color.

Observations Upon Natural Substances the Triboluminescence of Which May Be Classed with That of Fluorite

A number of specimens observed show, in general, the same characteristics as to TL and its relation to thermoluminescence as those exhibited by fluorite. Table IV gives a list of these substances. The materials in Table IV are classified with fluorite because they show TL in the natural state and stronger TL and also thermoluminescence of the same color after exposure to radium. They still show TL after they are heated in the blast and also tribothermoluminescence.

The following differences between the behavior of these substances and that of fluorite may be noted: Specimens 2, 3, and 5, Table IV, show much stronger TL after being heated in the blast than in the natural state. The TL and thermoluminescence resulting from exposure to radium are of different colors for specimens 2 and 3 from the colors of the natural specimens and of the same specimens when heated in the blast. It appears as if the luminescence of these specimens may be made up of two components, one of which is excited by radium, the other not.

Observations Upon Natural Substances Which Show Triboluminescence which Differs Essentially from that of Fluorite

A number of substances were observed which show TL differing essentially from that of fluorite. These are given in Table V. These materials all show TL in the natural state which is neither increased nor decreased in intensity by exposure to radium or subjection to high temperature such as that of the blast. All of these specimens are excited to fluorescence and phosphorescence by ultraviolet light from the mercury lamp. The TL is not changed by exposure to light and none of the specimens are tribothermoluminescent. The TL of sphalerite, specimens 1 and 2, is so strong that it can be easily seen in a light room. Particles crushed between glass plates emit strong light. Sphalerite is neither phosphorescent nor fluorescent after exposure to radium. Nelson[5] photographed the TL spectrum of sphalerite and found a band between 5700 and 6100A. He also photographed the spectrum of sparklets from a steel file on an emery wheel and found a band extending through the visible spectrum from 4300 to 6600A with a maximum at 5800A. This shows that the sparklets give off a thermal radiation and the TL is selective emission. Willemite is phosphorescent

[20] W. P. Headden, Am. J. Sci. **21**, 301 (1906).
[21] W. P. Headden, Proc. Col. Sci. Soc. **12**, 399 (1923).
[22] Purple fluorite from Weardale, specimen 7, Table I, after certain exposure to radium, shows green TL and purple low temperature thermoluminescence. This is due to an excess of Eu which dominates in the TL but not in the thermoluminescence. The green bands of Tb and Dy which dominate in the TL can also be seen in the spectrum of the low temperature thermoluminescence but they are dim compared with the Eu band.

and thermoluminescent after exposure to radium. The TL appears to be equally bright after the material has been heated and the thermoluminescence has been exhausted.

The TL shown in materials listed in Table V appears to have no relation to thermoluminescence; it does not depend upon previous excitation in the way in which phosphorescence and thermoluminescence do. The spectra of all the specimens observed is broad banded.

Artificially Prepared Substances Which Show Scintillations

A number of materials which show scintillations when exposed to alpha-rays from radium have similar characteristics to those of the specimens listed in Table V. These substances are:

1. Phosphorescent zinc sulphide
2. Calcium tungstate
3. Cadmium tungstate

They were kindly furnished by Dr. Karlik.

1. Zinc sulphide shows strong TL which is not reduced by heating. It is strongly excited to phosphorescence by light but this excitation appears to have no effect upon the TL. It has no tribothermoluminescence and exposure to radium appears to make no difference on the TL.

2. Two specimens of calcium tungstate were observed, a French preparation and a specimen from Heyden. Both showed strong TL after they were heated in the blast and very dim tribothermoluminescence. They were phosphorescent after exposure to radium, dimly thermoluminescent and the TL was somewhat stronger than before exposure to radium. The luminescence of calcium tungstate is in the blue end of the spectrum and does not look very bright to the eye.

3. A French specimen of cadmium tungstate was examined. It showed very strong green TL which was not reduced by the heat of the blast and was not changed by exposure to radium. The specimen showed no phosphorescence and no thermoluminescence after exposure to radium and showed no tribothermoluminescence. Under ultraviolet light it showed a dim yellowish-green fluorescence. Its characteristics are similar to those of sphalerite.

It has been observed by Lenard, Schmidt and Tomaschek[2] that because scintillations may be seen to a temperature of 350°C this phenomenon must be ascribed to the "dauer" centers. It is interesting to note that the TL observed in all the specimens examined which are known to show scintillations also gives evidence of centers that cannot be destroyed by heat.

Some Synthetic Materials Containing Manganese

Many substances which are excited to thermoluminescence by exposure to radium show TL exactly similar to the thermoluminescence. It appears that the energy emitted in the two phenomena must have its source in the same centers. Preparations containing manganese, similar to some of those described by Wiedemann and Schmidt[23] as strongly thermoluminescent after exposure to cathode rays, were kindly prepared by Dr. Rona. These preparations, listed

[23] Wiedemann and Schmidt, Ann. de Chemie et de Phys. **56**, 201 (1895).

Table V. *Natural materials showing triboluminescence unlike that of fluorite.*

		Natural Material		Exposed to Ultraviolet Light		Heated in Blast	
	Substance, Source	TL	Thermo-luminescence	TL	Phosphorescence	TL	Thermo-luminescence
1	Sphalerite, Horn Silver mine, Frisco, Utah	Gold, very strong	None	Gold, very strong	Green, yellow, very strong	Gold, very strong	None
2	Sphalerite, Franklin, New Jersey	Gold, very strong	None	Gold, very strong	Green, very strong	Gold, very strong	None
3	Willemite, Franklin, New Jersey	Green, strong	None	Green, strong	Green, strong	Green, strong	None

TABLE VI. *Synthetic materials which contain manganese.*

	Material	Heated in Blast		Heated in Blast, Exposed to Radium		Cathodo-luminescence
		TL	Tribo-thermol.	TL	Tribothermol.	
1	$MgSO_4$+1% $MnSO_4$	None	None	Red, strong	Red, strong	Dark red, strong
2	$CdSO_4$+1% $MnSO_4$	Weak	Fairly good	Yellow, strong	Yellow, strong	Yellow
3	Na_2SO_4+1% $MnSO_4$	None	None	Reddish-yellow, strong	Reddish-yellow, strong	Brownish-yellow
4	$CaCO_3$+1% $MnCO_3$*			Exposed to Radium Orange, weak	Orange, weak	

* $CaCO_3$+1 percent $MnCO_3$ could not be heated in the blast because it decomposes when subjected to high temperature.

in Table VI, after exposure to radium, all show TL which appears to be exactly similar to the thermoluminescence indicating that the energy absorbed from the radium may be emitted in either of the two ways. A number of chemically pure substances, $PbSO_4$, $CaCO_3$, and $CdSO_4$, were also found to show thermoluminescence and TL of the same color after exposure to radium. All the materials which were observed to show thermoluminescence after exposure to radium show TL of the same color.

GLASS

It is well known that glass is tl.[16] It is also thermoluminescent after exposure to radium. In order to investigate any possible relations between the two phenomena four specimens of glass were observed as follows:

1. Microscopic cover-glass colored brown by long exposure to radium showed a strong green thermoluminescence appearing at a low temperature and persisting a long time.

2. Microscopic cover-glass colored violet by long exposure to radium showed thermoluminescence not so strong as that of specimen 1, which comes up at a higher temperature and is less persistent.

3. Glass from Jena was strongly thermoluminescent after 86 hours exposure to radium. The emission came up at a low temperature but was not very persistent.

4. Thüringen glass was not so strongly thermoluminescent after 86 hours exposure to radium as specimen 3. A higher temperature was required for the emission and the effect was more persistent.

All the specimens of glass observed showed green TL after previous heating to redness and they showed the same TL unchanged after exposure to radium. It appears that exposure to radium makes glass thermoluminescent but has no effect upon the TL.

Tribothermoluminescence was observed in all the specimens in different intensities, 1 and 4 being stronger than 2 and 3. It appears that tribothermoluminescence is associated with the more persistent type of thermoluminescence because it also comes up at a high temperature and is very persistent.

SUGAR

Sugar, as is well known, is strongly tl showing the nitrogen bands in the spectrum as observed by Longchambon.[14] Sugar covered with xylol and ground in a mortar shows no TL. No tribothermoluminescence could be observed before the sugar burned.

SUMMARY

1. In the emission of TL there are three distinct factors one or all of which may take part on the TL of any given substance:

(*a*) Emission of TL which closely resembles phosphorescence and low temperature thermoluminescence. This comes from unstable, excited centers which may be activated by exposure to radium, x-rays and cathode rays.

(*b*) Emission which is characteristic of the material itself and is not dependent upon any

other excitation than mechanical breaking or crushing. This emission comes from stable, heat resistent centers.

(*c*) Emission which is due to an electric discharge. This discharge shows the nitrogen bands in air.

2. For the fluorites, synthetic materials containing CaF_2 and the rare earths, Table II, the calcites, Table III, and the materials listed in Table IV.

Triboluminescence is due to (*a*), (*b*) and (*c*) separately or together. In materials heated in the blast the TL is due to (*b*) and (*c*). In materials heated in the blast ground under liquids the TL is due to (*b*). For natural materials listed in Table V and for the artificially prepared substances which show scintillations TL is due to (*b*) and (*c*). For the synthetic materials contained in Table VI and for CaF_2+Mn and $CaSO_4$+Mn TL is due to (*a*).

3. Materials which show marked TL contain small amounts of impurities which serve as activaters as is the case in other luminescent substances. Certain impurities dominate and others are suppressed. The intensity of TL is affected by heat treatment.

4. The principle of essential identity[24] applies to triboluminescence as it does to other forms of luminescence. In many cases the spectrum of TL has been shown to be identical with that of phosphorescence and low temperature thermoluminescence excited by exposure to radium. The tl spectrum of fluorite characterized by the sharp bands of the rare earths makes comparison easy.

5. Substances which may be activated to relatively high temperature, persistent thermoluminescence, thermoluminescence by exposure to radium, cathode rays or x-rays show tribothermoluminescence.

The author wishes to express her sincere appreciation of the generous assistance given by the following persons who contributed specimens of the natural materials upon which the observations were made. These included Dr. Karl Przibram, Dr. H. Haberlandt, Mr. Wm. M. Lemcke, President D. M. Nelson, Ward's Natural Science Establishment and a source in Franklin, New Jersey.

[24] Nichols and Howes, Phys. Rev. **27**, 803 (1926).

Note on Illumination Characteristics of the Camera Lens

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(Received May 19, 1937)

ONE important feature of the geometrical optics of the camera seems to be somewhat neglected in the literature. The illumination received on various sections of a camera film depends upon the location of the sections with respect to the optic axis of the camera. This

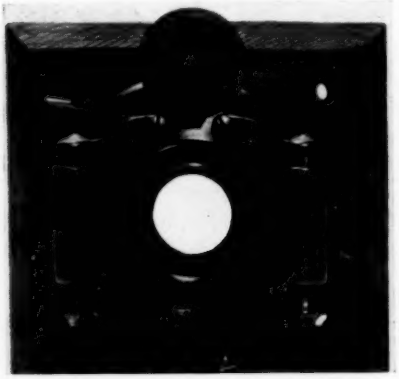

FIG. 1. The effective aperture of this lens, when viewed from a point on the optic axis, is circular in form.

might seem like a self-evident fact to any student of optics, but there have recently appeared articles[1, 2] on photographic photometry in which the author seemingly assumed that all parts of the film would be equally exposed when photographing equally bright objects occupying various parts of the field.

Let us assume as the simplest possible case, that the lens is thin, and that the thickness of the lens and mounting may be neglected. Viewed from the central point of the film the lens would appear as a circular aperture of area A_0, and the brightness of the lens would be equal to that of the point of the external field being viewed, except for the reflection and transmission losses in the glass. Viewed from some other point at an angle of θ degrees from the axis of the camera, the angular size of the lens is less, and it now appears foreshortened into an ellipse whose minor axis is less than the major axis by the factor $\cos\theta$, and the apparent area becomes $A_0 \cos\theta$. The intensity of the light from the lens, when we consider it as the source of light, is therefore, for a uniformly bright external field,

$$I_\theta = I_0 \cos\theta \text{ candles},$$

where I_0 is the intensity along the axis and I_θ is the intensity at an angle of θ degrees from the axis. If the focal distance along the axis is F, then the illumination on the central point of the film is

$$E_0 = (I_0/F^2) \text{ ft.-c.}$$

At a point θ degrees from the axis, the distance from lens to film is increased by the factor $1/\cos\theta$ and the light strikes the film at the angle θ. Taking these two additional factors into account, the illumination at the angle θ is

$$E_\theta = (I_0/F^2) \cos^4\theta \text{ ft.-c.}$$

It therefore is apparent that the illumination from an ideal thin lens (or pinhole aperture) varies as the fourth power of $\cos\theta$. One conventional size of film has a half-width of $0.33F$

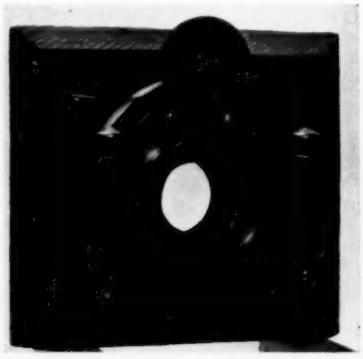

FIG. 2. This is the same lens as in Fig. 1, but viewed from an angle of 20 degrees and from a slightly greater distance. The effective aperture is the same when viewed from the film side of the lens.

[1] J. M. Waldram, "A Contouring Density Comparator," J. Sci. Inst. (British) **13**, 352 (1936).
[2] A. Bloch, "Measurement of Nonuniform Brightness by Photographic Photometry," J. Sci. Inst. (British) **13**, 358 (1936).

FIG. 3. At 40 degrees from the axis the effective area of lens is nearly zero and this angle marks the extreme width of field possible with this lens.

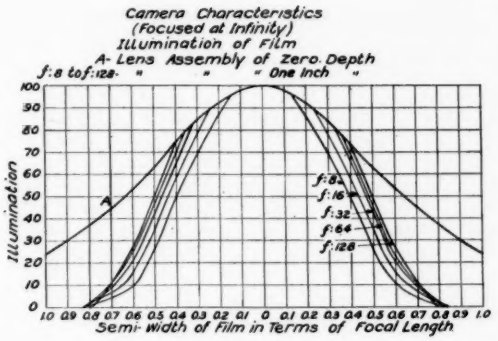

FIG. 4.

and a half-length of $0.40F$. The dimensions correspond to angles of 18 degrees and 22 degrees. The reduction of illumination, as computed from the above equation, is about 18 percent across the narrow way of the film, and 26 percent at the center of the ends of the film. If the extreme corners are considered, the illumination will be found to be only 60 percent of the central illumination.

In the case of wide angle lenses θ may be 45 degrees, and then the edge illumination falls to 25 percent of the central illumination.

So far we have considered only the ideal case of a thin lens. Actual camera lenses have depth between the front and rear elements, and this depth has a strong influence on the amount of transmitted light. In Fig. 1 is shown a front view of a camera lens set at $f:8$. A rear view from the center of the film would show the same size and form of effective lens area, which is entirely determined by the setting of the diaphragm. In Fig. 2, the same lens is seen from an angle of 20 degrees and from a distance corresponding to the 20 degree point on the film. The active lens area is outlined on the left edge by the front lens mount, on top and bottom by the iris diaphragm, and on the right by the mount of the rear lens. It can be seen from the photograph that the iris diaphragm now plays a minor part in determining the area of lens that is effective at 20 degrees from the axis.

At 40 degrees from the axis the transmission is practically zero, as shown by Fig. 3, and here the diaphragm plays no part whatever in determining the effective aperture.

The distribution of light over the film, using the above lens, is illustrated in Fig. 4. The top curve, marked A, is computed for a thin lens, and applies to any lens when only the diaphragm is effective in fixing the effective area of the lens. The curve marked $f:8$ applies to this particular lens when set at a speed of $f:8$. The other curves are for other speeds down to $f:128$. Going back to the standard film size of $0.33F$ by $0.40F$, these curves show that for a speed of $f:8$ the edge illuminations will fall to 64 percent and 50 percent, while the corners will receive only 20 percent of the light falling on the center of the film.

These data apply to this lens only, and for each design of lens assembly there will be a different group of curves for various iris settings, but they will all start from curve A, which is perfectly general.

The variations here pointed out are not of particular interest to the casual photographer, but if photographic photometry is attempted it would seem that the camera is severely handicapped by its own geometry. This handicap can be removed only by a study of the lens assembly, and for really precise work the study should of course be more thorough than the elementary outline here given.

Analysis of Doubly Refracting Materials with Circularly and Elliptically Polarized Light

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INTRODUCTION

WHEN a doubly refracting crystal plate is examined at normal incidence between "crossed Nicols," two types of interference bands are observed. These are the fringes (isochromatics) and the brushes (isoclinics). The former depend upon the relative retardation of the ordinary and extraordinary rays and the latter depend upon the orientations of the polarizing axes of the crystal. It is often desirable to eliminate the isoclinics in order to better view the retardation fringes. This is usually accomplished by introducing, between the specimen and the polarizers, a pair of retardation plates with polarizing axes oriented at 45° with respect to the vibration directions of the polarizers. If the retardation plates are true quarter-wave plates for the monochromatic light source, it is well known that the isoclinics are completely removed, while the retardation fringes remain unaltered.

It is the purpose of this paper to show that, when the retardation plates are not accurately quarter-wave plates, not only are the isoclinics not completely removed but also the retardation fringes are not necessarily unchanged. These conclusions have been reached through an analysis of the equations governing the intensity

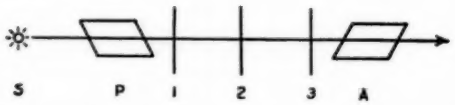

FIG. 1. Schematic diagram of polarizing apparatus. S=monochromatic light source; P=polarizer; 1 and 3 =retardation plates; 2=specimen; A=analyzer.

of monochromatic light transmitted through a polarizer, three doubly refracting plates and an analyzer. It appears that the retardation fringes are unaltered if the two retardation plates are identical, regardless of the retardation introduced by each one; but, if the two plates are not identical, the fringes are displaced by an amount which depends, in part, upon the difference in retardations of the two plates.

This phenomenon is important, for example, in photoelastic work in which the magnitude of the stress at a point is calculated from the order of the fringe appearing at that point. The displacement of a fringe, caused by inaccuracies in the retardation plates, introduces an error, in the stress, which may be calculated from the equations given below, provided that the retardations of the two retardation plates are known.

INTENSITY EQUATIONS

Figure 1 shows, schematically, the optical arrangement under consideration. Light from a monochromatic source, S, passes successively at normal incidence through a polarizer, P, a retardation plate, 1, the specimen, 2, a second retardation plate, 3, and an analyzer A. Fig. 2 depicts the orientations of the polarizing axes as seen by an observer viewing the oncoming light from the analyzer end of the system. P is the vibration direction of the light transmitted through the polarizer; ρ_1, ρ_2, ρ_3 are the orientations of the fast axes of the first retardation plate, the specimen, and the second retardation plate, respectively, measured positive counter clockwise from P; and θ is the orientation of the vibration direction of the analyzer, referred to P.

In the equations which follow, I is the intensity of light transmitted through the system; I_0 is the intensity transmitted through the system when $\rho_1=\rho_2=\rho_3=\theta=0$ and δ_1, δ_2, δ_3 are the relative phase retardations introduced by the first retardation plate, the specimen and the second retardation plate, respectively. For arbitrary values of all the orientations and retardations the equation which governs the intensity is[1]

[1] For a method of deriving formulas such as Eq. (1), see G. Szivessy, *Handbuch der Physik*, Vol. 19, p. 924.

$$\frac{2I}{I_0}=\sin 2(\theta-\rho_3)\left\{\begin{array}{l}-\cos 2\rho_1 \cos 2(\rho_2-\rho_1) \sin 2(\rho_3-\rho_2) \cos \delta_3 \\ +\sin 2\rho_1 \sin 2(\rho_2-\rho_1) \sin 2(\rho_3-\rho_2) \cos \delta_3 \cos \delta_1 \\ -\cos 2\rho_1 \sin 2(\rho_2-\rho_1) \cos 2(\rho_3-\rho_2) \cos \delta_3 \cos \delta_2 \\ +\cos 2\rho_1 \sin 2(\rho_2-\rho_1) \sin \delta_3 \sin \delta_2 \\ -\sin 2\rho_1 \cos 2(\rho_2-\rho_1) \cos 2(\rho_3-\rho_2) \cos \delta_1 \cos \delta_2 \cos \delta_3 \\ +\sin 2\rho_1 \cos 2(\rho_2-\rho_1) \sin \delta_2 \sin \delta_3 \cos \delta_1 \\ +\sin 2\rho_1 \cos 2(\rho_2-\rho_1) \cos \delta_3 \sin \delta_2 \sin \delta_1 \\ +\sin 2\rho_1 \sin \delta_3 \cos \delta_2 \sin \delta_1\end{array}\right\}$$

$$+\cos^2(\theta-\rho_3)\left\{\begin{array}{l}2\cos^2\rho_1\cos^2(\rho_2-\rho_1)\cos^2(\rho_3-\rho_2) \\ +2\cos^2\rho_1\sin^2(\rho_2-\rho_1)\sin^2(\rho_3-\rho_2) \\ +2\sin^2\rho_1\sin^2(\rho_2-\rho_1)\cos^2(\rho_3-\rho_2) \\ +2\sin^2\rho_1\cos^2(\rho_2-\rho_1)\sin^2(\rho_3-\rho_2) \\ -\sin 2\rho_1 \sin 2(\rho_2-\rho_1) \cos 2(\rho_3-\rho_2) \cos \delta_1 \\ -\cos 2\rho_1 \sin 2(\rho_2-\rho_1) \sin 2(\rho_3-\rho_2) \cos \delta_2 \\ -\sin 2\rho_1 \cos 2(\rho_2-\rho_1) \sin 2(\rho_3-\rho_2) \cos \delta_1 \cos \delta_2 \\ +\sin 2\rho_1 \sin 2(\rho_3-\rho_2) \sin \delta_1 \sin \delta_2\end{array}\right\}. \qquad (1)$$

$$+\sin^2(\theta-\rho_3)\left\{\begin{array}{l}2\cos^2\rho_1\cos^2(\rho_2-\rho_1)\sin^2(\rho_3-\rho_2) \\ +2\cos^2\rho_1\sin^2(\rho_2-\rho_1)\cos^2(\rho_3-\rho_2) \\ +2\sin^2\rho_1\sin^2(\rho_2-\rho_1)\sin^2(\rho_3-\rho_2) \\ +2\sin^2\rho_1\cos^2(\rho_2-\rho_1)\cos^2(\rho_3-\rho_2) \\ +\sin 2\rho_1 \sin 2(\rho_2-\rho_1) \cos 2(\rho_3-\rho_2) \cos \delta_1 \\ +\cos 2\rho_1 \sin 2(\rho_2-\rho_1) \sin 2(\rho_3-\rho_2) \cos \delta_2 \\ +\sin 2\rho_1 \cos 2(\rho_2-\rho_1) \sin 2(\rho_3-\rho_2) \cos \delta_1 \cos \delta_2 \\ -\sin 2\rho_1 \sin 2(\rho_3-\rho_2) \sin \delta_1 \sin \delta_2\end{array}\right\}$$

Plane Polariscope

If the specimen is viewed simply between crossed Nicols, the optical system is known as a plane polariscope. The intensity of light transmitted through such a system is obtained by substituting, in Eq. (1), $\delta_1=\delta_3=0$ and $\theta=\pi/2$. Then

$$I=I_0 \sin^2(\delta_2/2) \sin^2 2\rho_2. \qquad (2)$$

Extinction of light is observed when

$$\delta_2=2n\pi, \quad (n=\pm1, \pm2, \pm3, \cdots) \qquad (3)$$

and also when

$$\rho_2=n\pi/2. \qquad (4)$$

Dark bands formed according to Eq. (3) are the retardation fringes and those governed by Eq. (4) are the isoclinics.

Circular Polariscope

To remove the isoclinics, quarter-wave retardations are assigned to plates 1 and 3 and the polarizing axes of the two plates are oriented at angles of 45° to the vibration directions of P and A. The optical arrangement is known as a circular polariscope. In Eq. (1), $\delta_1=\delta_3=\pi/2$, $\rho_1=\pi/4$, $\rho_3=3\pi/4$, $\theta=\pi/2$. The intensity of monochromatic light transmitted through a circular polariscope is, then, given by

$$I=I_0 \sin^2(\delta_2/2). \qquad (5)$$

The intensity is accordingly independent of the orientation (ρ_2) of the polarizing axes in the specimen, so that the isoclinics do not appear; but the retardation fringes again occur according to Eq. (3).

Balanced Polariscope

Suppose, now, that the two retardation plates are not true quarter-wave plates for the monochromatic source, but that they are, nevertheless, identical. Substituting in Eq. (1) the values $\delta_1=\delta_3=\delta$ and $\rho_1=\pi/4$, $\rho_3=3\pi/4$, $\theta=\pi/2$, there results

$$I=I_0 \sin^2(\delta_2/2)(1-\cos^2 2\rho_2 \cos^2\delta). \qquad (6)$$

The quantity in parentheses in Eq. (6) shows that, in this case, the isoclinics are not completely removed. The proportion which remains depends upon the relative retardation of each of the two retardation plates and follows a cosine-squared law. When $\delta \to 0$ the isoclinics are completely restored and Eq. (6) becomes identical with Eq. (2). When $\delta \to \pi/2$ (quarter-wave plates) the isoclinics are completely removed and Eq. (6) approaches Eq. (5).

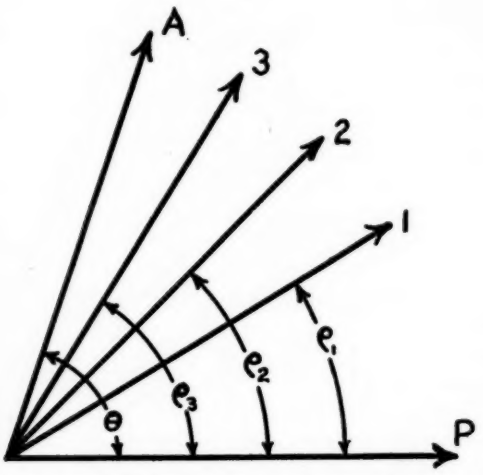

FIG. 2. Angular relations of vibration direction of analyzer A, fast polarizing axes of retardation plates (1, 3) and specimen (2); all referred to vibration direction of polarizer (P).

No matter what value is assigned to δ, the retardation fringes always occur according to Eq. (3) since δ_2 appears in Eq. (6) only in the form of the multiplying factor $\sin^2(\delta_2/2)$ just as in Eq. (2).

The relation expressed by Eq. (6) was used by R. V. Baud[2] to show that, with a *white* light surce, the isochromatic fringes vary with the orientation (ρ_2) of the polarizing axes in the specimen.

UNBALANCED POLARISCOPE

If the two retardation plates are not identical, it appears that even with monochromatic light, the fringe pattern is a function of the orientation of the polarizing axes in the specimen. Furthermore, for a given inclination of the polarizing axes the fringe pattern observed also depends upon the relative retardations of the two retardation plates.

The intensity equation governing this unbalanced case is obtained by substituting $\rho_1=\pi/4$, $\rho_3=3\pi/4$, $\theta=\pi/2$ in Eq. (1), and there results:

$$I=I_0\left\{\sin^2\frac{\delta_2}{2}[\cos(\delta_3-\delta_1)-\cos^2 2\rho_2\cos\delta_1\cos\delta_3]\right.$$
$$\left.+\sin^2\frac{\delta_3-\delta_1}{2}-\tfrac{1}{2}\sin\delta_2\sin 2\rho_2\sin(\delta_3-\delta_1)\right\}. \quad (7)$$

The appearance of the term $\sin\delta_2$ in Eq. (7) indicates that, when $\delta_3 \neq \delta_1$, the fringes no longer occur according to Eq. (3). There is then observed a displacement of the fringe pattern which is further complicated by the presence of the multiplying factor $\sin 2\rho_2$. Consequently, there is a complex distortion of the fringe pattern when both the relative retardation and the inclination of the polarizing axes are not uniform throughout the specimen, as is the case in a photoelastic model.

Instead of fringes appearing according to Eq. (3), maximum and minimum intensities will be observed in an unbalanced polariscope when

$$\tan\delta_2=\frac{\sin 2\rho_2\sin(\delta_3-\delta_1)}{\cos(\delta_3-\delta_1)-\cos^2 2\rho_2\cos\delta_1\cos\delta_3}. \quad (8)$$

For a given pair of retardation plates, the maximum error in calculating relative retardation from fringe order occurs when the polarizing axes in the specimen are at 45° to the vibration direction of the polarizer. Eq. (8) then reduces to

$$\tan\delta_2=\pm\tan(\delta_3-\delta_1)$$

and therefore a dark fringe appears when

$$\delta_2=2n\pi\pm(\delta_3-\delta_1). \quad (9)$$

The upper sign is associated with positive values of $\sin 2\rho_2$ and the lower sign with negative values of $\sin 2\rho_2$.

CONCLUSIONS

(1) Equation (7) shows that the fringe pattern is distorted in an optically unbalanced polari-

[2] R. V. Baud, J. O. S. A. **20**, 386 (1930), and **21**, 119 (1931).

scope. The distortion, for a given wave-length, is a function of the orientation of the specimen and the phase retardations of the two retardation plates.

(2) The magnitude of the error introduced by the optical unbalance (with monochromatic light) may be calculated from Eq. (8).

(3) Equation (9) shows that the *maximum* error (in calculating relative retardation from fringe order) that can occur in an optically unbalanced polariscope is given by the difference between the phase retardations of the two retardation plates.

(4) If the retardation plates in a polariscope are different from quarter-wave plates, but are nevertheless identical, it follows from Eq. (6) that the fringe pattern is unaffected. The only effect will be incomplete removal of isoclinics.

(5) Equation (6) also shows that if a monochromatic light source of wave-length other than that for which the instrument is designed is substituted in a balanced polariscope, the fringe pattern will remain unchanged.

A Simple Brightness Meter

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(Received June 7, 1937)

BRIGHTNESS and brightness contrasts are the most generally important among the fundamental factors affecting visibility. This is recognized in research, but not in everyday seeing as is evidenced by the lack of available brightness meters. Luckiesh and Taylor[1] have recently described a refined brightness meter having the enormous range of 25,000,000 to 1 with the accuracy and convenience desirable for a vast variety of refined measurements wherever absolute brightness is important. The instrument described in the present note is illustrative of extreme simplicity with some sacrifice in convenience. Its range can be as great as is necessary. It measures relative brightness and brightness contrasts and depends upon any available footcandle meter to convert relative brightness into absolute brightness. The simple form described involves a fundamental principle which can take many physical forms besides the one actually illustrated.

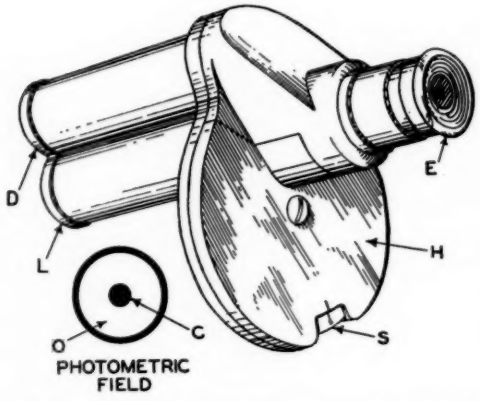

FIG. 1. A simple brightness meter for measuring brightness contrasts. Its determinations of relative brightness are readily convertible into absolute brightness with the aid of any footcandle meter by measuring the footcandles on the diffusing glass, *D*.

One tube of the device has a lens *L* in the end which, in combination with the adjustable eyepiece *E*, is practically a telescope of universal focus. The instrument is so sighted that the image of the object whose brightness *C* to be measured is seen in the central portion of the photometric field surrounded by a mirrored portion. In the end of the other tube is a perfectly diffusing translucent glass. Its brightness *O* is proportional to the footcandles upon it and its brightness is seen in the mirrored portion of the photometric field. Thus *O* provides the comparison brightness. The brightness of the object focused is varied by means of a circular gradient of extensive range in transmission factor which is enclosed in the housing *H*. Thus a setting is made and the relative brightness of the object is read from the scale *S*.

Suppose the relative brightnesses of the envelope of diffusing glass of a common office lighting fixture and that of the ceiling surrounding it are to be determined. The foregoing procedure is applied first to the bright portion of the fixture and then to the adjacent ceiling. In both cases the illumination in footcandles upon the diffusing glass *D*, and hence its brightness, remain sensibly the same. Thus *D* is a constant comparison field for the determination of relative brightnesses of the fixture and ceiling and the brightness contrast, which is an important factor in visibility and comfort. Inasmuch as the brightness of *D* depends upon its illumination, a simple measurement of the footcandles received by *D* provides a means of conversion (with the aid of a predetermined constant for the instrument) of relative into absolute brightness. With the increasing use and availability of inexpensive footcandle meters such a simple brightness meter should eventually serve many practical purposes.

The actual range in brightness obtainable by the gradient alone is of the order of 2000 to 1. Reducing and multiplying screens can be inserted to extend the range. These are incor-

[1] Matthew Luckiesh and A. H. Taylor, "A Brightness Meter," J. O. S. A. **27**, 132 (1937).

porated in the instrument on the side not shown in the illustration. If screens having transmission factors of ten percent and one percent are arranged to be readily inserted on either side of the photometric field, the range is extended to many millions from the lowest brightness that the visual sense can detect with ease.

Besides the many obvious applications in the measurement of brightness, this instrument by itself is useful in the study of brightness contrasts in the field of lighting because they have much influence upon comfort in seeing and some influence upon visibility. Quality of lighting, which includes chiefly the distribution of brightness in the visual field, should be studied by lighting specialists along with brightness. Our development of this device has been largely inspired by this need. Footcandle measurements are common and necessary, but they have overshadowed measurements of brightness and brightness contrast entirely too much in practice and sometimes in theory.[2]

[2] Matthew Luckiesh and Frank K. Moss, *The Science of Seeing* (D. Van Nostrand Co., New York, 1937).

Projective Transformations of I. C. I. Color Specifications

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(Received May 25, 1937)

The use of the I. C. I. 1931 Standard Observer and Coordinate System for Colorimetry for all general color specifications and calculations is urged for the elimination of confusion in the field of colorimetry. Methods for constructing other coordinate systems are exhibited, and interpretations are given for the eight independent constants in the equations for transformation from the I. C. I. to other coordinate systems. Expressions are derived for the luminosity coefficients in terms of the constants of the equations for the transformation of coordinates. The possibility of assigning luminosity coefficients without restricting the shape of the spectrum locus is discussed, and an example is given. The assignment of luminosity coefficients interferes with the assignment of the shape of the spectrum locus only in the case of three equal luminosity coefficients. In general, two of the eight degrees of freedom which are available in the defining equations of a trichromatic coordinate system are eliminated by the assignment of the luminosity coefficients. Except in the case of three equal luminosity coefficients, four of the six remaining degrees of freedom are eliminated when the shape of the spectrum locus is established. One additional degree of freedom is eliminated when the size of the spectrum locus is assigned. The final degree of freedom is most profitably used for the purpose of simplifying the transformation equations. The entire discussion is based on well-known principles of transformation of color mixture data and projective geometry; no assumptions other than those underlying standard trichromatic color specification are made, and no conclusions of physiological or psychological import are justified.

RECENT articles by D. B. Judd[1,2] have suggested the possibility of constructing chromaticity diagrams in which approximately equal distances separate the points representing all pairs of equally bright colors which are just noticeably different under certain specified conditions of observation. This possibility has reawakened interest[3,4] in methods for the transformation of trichromatic color specifications and in the projective properties of color mixture diagrams. These subjects have been discussed by H. E. Ives,[5] J. Guild,[6] D. B. Judd,[7] and several others. The following discussion will not contain any essentially new principles, but will describe practical methods which have been found useful in applying Judd's device for simplifying the representation of tolerance data in color specifications.

A. The I. C. I. System Urged for Universal Use as System of Original Specification

The only type of modification of color specifications that appears to be of current interest is the linear combination type of transformation of the I. C. I.[8] color mixture data, and the

[1] D. B. Judd, "A Maxwell Triangle Yielding Uniform Chromaticity Scales," J. O. S. A. **25**, 24 (1935).

[2] D. B. Judd, "Estimation of Chromaticity Differences and Nearest Color Temperature on the Standard 1931 I. C. I. Colorimetric Coordinate System," J. O. S. A. **26**, 421 (1936).

[3] F. C. Breckenridge, and W. R. Schaub, "Rectangular Uniform-Chromaticity-Scale Coordinates," Abstract No. 14, J. O. S. A. **27**, 226 (1937).

[4] R. H. Sinden, "Further Search for the Ideal Color System, I," J. O. S. A. **27**, 124 (1937).

[5] H. E. Ives, "The Transformation of Color Mixture Equations from One System to Another," J. Frank. Inst. **180**, 673 (1915); also "The Transformation of Color Mixture Equations from One System to Another, II. Graphical Aids," J. Frank. Inst. **195**, 23 (1923).

[6] J. Guild, "The Transformation of Trichromatic Mixture Data: Algebraic Method," Trans. Opt. Soc. **26**, 95 (1924); also "The Geometrical Solution of Color Mixture Problems," Trans. Opt. Soc. **26**, 139 (1924).

[7] D. B. Judd, "Reduction of Data on Mixture of Color Stimuli," Nat. Bur. Stand. J. Research **4**, 515 (1930).

[8] Proc. 8th Session, Commission International de l'Eclairage, Cambridge, 1931. Note that the initials, C. I. E., which refer to the French form of the title are sometimes used as an abbreviation instead of the initials, I. C. I. Also, T. Smith, and J. Guild, "The C. I. E. Colorimetric Standards and Their Use," Trans. Opt. Soc. **33**, 73 (1931). Also, D. B. Judd, "The I. C. I. Standard Observer and Coordinate System for Colorimetry," J. O. S. A. **23**, 359 (1933). The possibility of future changes in the international system of colorimetric specification cannot be denied. The fact that such changes would render obsolescent the tremendous mass of colorimetric data which is being accumulated by many laboratories in this country and abroad at an accelerating rate now totaling several hundred samples each working day appears to be creating an inertia which will tend to prevent any changes which are not necessitated by the discovery of extremely serious errors in the 1931 recommendations. Evidence of errors of such a serious nature has not as yet been reported by any worker. The particular unitary stimuli in which the I. C. I. specifications are expressed have no peculiar merits, other than convenience for computation, which would lead to their adoption in preference to the ninefold infinity of other conceivable choices of unitary stimuli. They are peculiarly important only in that they are being used widely as bases for colorimetric specifications.

consequent projective type of transformation of the I. C. I. chromaticity diagram. In the following discussion, the color mixture functions and the trichromatic coordinate system adopted in 1931 by the International Commission on Illumination will be assumed as the system of original specification from which all transformed specifications will be derived. The present paper will demonstrate the simplicity with which I. C. I. data can be transformed for special purposes. Useful as these transformed specifications may prove to be, instances will occur frequently in which the specifications in some other system will be desired. The greatest flexibility of colorimetric specification will be attained with the greatest ultimate economy of computation, if the I. C. I. specifications are always available, and if each new system of specification designed for special purposes is derived as a simple transformation of the I. C. I. specification.

The computations of I. C. I. specifications from spectrophotometric data have been greatly expedited by the construction of tables of selected ordinates.[9] A tremendous amount of essentially unnecessary labor would be expended if such tables were constructed for each of the numerous modifications of the coordinate system which will be proposed for special purposes by various workers in the near future. Furthermore, for each new system, in which original computations might then be made, it would be necessary to derive separate sets of transformation equations for the conversion of the data to and from each of the other existing systems. If, as outlined above, the I. C. I. specifications are always determined first for every sample, then only one new set of transformation equations will be necessary for the transformation of the available data into any new coordinate system. It is the author's opinion that avoiding confusion which would result from the use of many different bases for color specification is the responsibility of and to the advantage of every worker in the field of colorimetry.

The second category of reasons for the adoption of the I. C. I. system as the system of original specification for which the following derivations and methods are valid concerns the simplifications which are introduced by the convenient luminosity coefficients, 0–1–0, of the I. C. I. unitary stimuli and by the location of the equal energy point at the center of the I. C. I. diagram. The following analysis is valid only for transformations from systems having these characteristics.

In the following discussion, the symbols for the I. C. I. data which were introduced by the I. C. I. report will be adopted:

The values X, Y, Z are called the tristimulus values of the sample and are the amounts of three I. C. I. unitary color stimuli which must be mixed optically in order to match the color of the sample.

The values $\bar{x}_\lambda$, $\bar{y}_\lambda$, $\bar{z}_\lambda$ are the tristimulus values of a unit amount of radiant energy of wavelength λ. These are often called color mixture functions, or distribution functions.

The values x, y are the coordinates of the point representing the chromaticity of the sample in the I. C. I. rectangular coordinate system, and by definition

$$x=X/(X+Y+Z), \quad y=Y/(X+Y+Z).$$

The symbol m has been used previously[9, 10] as a convenient symbol for the denominators of these fractions: $m=X+Y+Z$. The location in the I. C. I. chromaticity diagram of the point representing any additive mixture of colors is the center of gravity of the points representing the component colors, when each of these points is weighted with the corresponding value of m.

B. Equations for Transformation of I. C. I. Specifications into Other Systems

The tristimulus values, U, V, W, of any sample expressed in terms of any new set of unitary stimuli can be computed from the I. C. I. tristimulus values, if the nine constants in the following equations are known.

$$\begin{aligned} U&=a_1X+a_2Y+a_3Z, \\ V&=a_4X+a_5Y+a_6Z, \\ W&=a_7X+a_8Y+a_9Z. \end{aligned} \tag{1}$$

It is evident that the constants, a_1, a_4, a_7, are, respectively, the values of U, V, W for the I. C. I. unitary stimulus, for which $X=1.0$, $Y=0.0$, and

[9] A. C. Hardy, *Handbook of Colorimetry* (Technology Press, Cambridge, Mass., 1936).

[10] D. L. MacAdam, "Theory of Maximum Visual Efficiency of Colored Materials," J. O. S. A. **25**, 249 (1935).

$Z=0.0$. Similarly, a_2, a_5, a_8 are the values of U, V, W for the I. C. I. unitary stimulus, for which $X=0.0$, $Y=1.0$, and $Z=0.0$. Finally, a_3, a_6, a_9

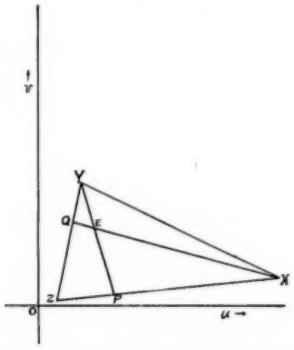

FIG. 1. A representation of the I. C. I. coordinate system in a new coordinate system.

are the values of U, V, W for the I. C. I. unitary stimulus, for which $X=0.0$, $Y=0.0$, $Z=1.0$.

It can easily be shown that U, V, W are also given by the following equations in terms of x, y, and m:

$$\begin{aligned} U&=m[(a_1-a_3)x+(a_2-a_3)y+a_3], \\ V&=m[(a_4-a_6)x+(a_5-a_6)y+a_6], \\ W&=m[(a_7-a_9)x+(a_8-a_9)y+a_9]. \end{aligned} \tag{2}$$

The coordinates u, v, in the new chromaticity diagram are defined analogously to the I. C. I. coordinates:

$$u=U/(U+V+W), \quad v=V/(U+V+W).$$

If m' is defined as the sum of the new tristimulus values, $m'=U+V+W$, then

$$u=U/m', \quad v=V/m'.$$

The transformation equations for the coordinates, u, v, of the sample in the new system, in terms of the coordinates in the old system result from the substitution of Eqs. (2) in these definitions, and involve eight independent constants.

$$\begin{aligned} u&=(c_1x+c_2y+c_3)/(c_7x+c_8y+1), \\ v&=(c_4x+c_5y+c_6)/(c_7x+c_8y+1). \end{aligned} \tag{3}$$

Although it will be desirable later to assign definite units to U, V, W, in order to secure desired absolute values for the luminosity coefficients, only the relative values are important for the following analysis of the significance of the constants, $c_1 \cdots c_8$. Consequently, the condition that $a_3+a_6+a_9=1.0$ can be imposed temporarily. With this condition,

$$\begin{aligned} &c_1=a_1-a_3, \quad c_2=a_2-a_3, \quad c_3=a_3, \\ &c_4=a_4-a_6, \quad c_5=a_5-a_6, \quad c_6=a_6, \\ &c_7=a_1+a_4+a_7-1, \quad c_8=a_2+a_5+a_8-1. \end{aligned} \tag{3a}$$

C. Significance of Constants in Equations for Transformation of Coordinates

A consideration of the definition of m' and of the significance of the constants $a_1 \cdots a_9$ of Eqs. (1), as discussed above, shows that:

$$m'=m(c_7x+c_8y+1). \tag{4}$$

If m'_X, m'_Y, m'_Z designate the values of m' for the three I. C. I. unitary stimuli, then

$$\begin{aligned} m'_X&=a_1+a_4+a_7=c_7+1, \\ m'_Y&=a_2+a_5+a_8=c_8+1, \\ m'_Z&=a_3+a_6+a_9=1. \end{aligned} \tag{4a}$$

The constants $c_1 \cdots c_8$ are uniquely determined by the assignment of the location in the new coordinate system of any four chromaticities no one of which can be matched by additive mixture of two of the others. The assignment of the locations of the three I. C. I. unitary stimuli and of the point E, representing the chromaticity of a uniform distribution of energy throughout the visible spectrum, is accomplished very conveniently in terms of the constants, $c_1 \cdots c_8$.

Let the coordinates of the I. C. I. unitary stimuli in the new coordinate system be denoted by u_X, v_X, u_Y, v_Y, u_Z, v_Z. In Fig. 1, the locations of the I. C. I. unitary stimuli in the new coordinate system are indicated by the points X, Y, Z. The location of the equal energy point, E, in the new coordinate system is then determined by the values of m'_X and m'_Y which are assigned to the I. C. I. unitary stimuli in the new system. The center of gravity principle applied to Fig. 1 reveals that the line through the points Y and E divides the line ZX in the ratio $ZP/PX=m_X'=c_7+1$, and the line through the points X and E divides the line ZY in the ratio $ZQ/QY=m_Y'=c_8+1$. Therefore, the assignment of the location of the equal energy point, E, relative to the points X, Y, Z, representing the I. C. I. unitary stimuli in the new coordinate system, determines the constants c_7 and c_8. The remaining six

constants are determined by substitution of the x and y coordinates of the I. C. I. unitary stimuli in Eqs. (3).

$$\begin{aligned} c_7&=ZP/PX-1=m'_X-1,\\ c_8&=ZQ/QY-1=m'_Y-1,\\ c_3&=u_Z, \qquad c_6=v_Z,\\ c_1&=u_Xm'_X-u_Z, \quad c_4=v_Xm'_X-v_Z,\\ c_2&=u_Ym'_Y-u_Z, \quad c_5=v_Ym'_Y-v_Z. \end{aligned} \tag{5}$$

Only three of these eight degrees of freedom are necessary in order to fix the shape and size of the triangle, XYZ, formed by the lines joining the points representing the I. C. I. unitary stimuli. For instance, the lengths of the segments ZX and ZY and the cosine of the angle XZY are sufficient to determine the size and shape of this triangle. These three quantities, together with the two ratios, ZP/PX and ZQ/QY, which locate the white point, fix the size and shape of the spectrum locus. The methods outlined by Judd,[1] and Sinden,[4] can be used in the search for useful modifications of the size and shape of the spectrum locus. The three remaining degrees of freedom can be used in the attainment of additional desirable features of the coordinate system. For instance, convenient ratios of the luminosity coefficients can be obtained, and the equations of transformation from the I. C. I. specifications can be thrown into very simple forms.

D. Luminosity Coefficients of Transformed Tristimulus Values

The luminosity coefficients, L_u, L_v, L_w, of the tristimulus values, U, V, W, are defined as the coefficients of U, V, W in the explicit solution of Eqs. (1) for the I. C. I. tristimulus value, Y, which is proportional to the luminosity of any sample color:

$$Y=L_uU+L_vV+L_wW.$$

Equations (1) can be solved for Y, and after substitution of the values of $a_1\cdots a_9$ which can be obtained from Eqs. (3a), (4a), and (5), the luminosity coefficients are found to be:

$$\begin{aligned} L_w&=(u_Zv_X-u_Xv_Z)/D,\\ L_u&=(v_z-v_x)/D+L_w,\\ L_v&=(u_X-u_Z)/D+L_w, \end{aligned} \tag{6}$$

where

$$D=(m'_Y+1)[v_X(u_Z-u_Y)+v_Y(u_X-u_Z)+v_Z(u_Y-u_X)].$$

The denominator, D, will be different from zero for all nondegenerate transformations. Consequently, L_w will be zero, if $u_Zv_X-u_Xv_Z=0$. If L_w is zero, then L_u will also be zero if $v_X=v_Z=0$. Under these circumstances, $L_v=1/(m_Y'+1)v_Y$. The luminosity coefficient, L_v, can now be made equal to 1.0 without disturbing any of the adjustments previously made, if all of the coefficients of Eqs. (1) are multiplied by the numerical value of $(m_Y'+1)\cdot v_Y$. This abandons the previously imposed condition that $a_3+a_6+a_9=1$, which was introduced for the sole purpose of simplifying the analysis.

The condition, $v_X=v_Z=0$, expresses the well-known principle that the luminosity coefficients for two of any set of unitary stimuli will be zero, if both of these unitary stimuli lie on the straight line, called the alychne, which passes through the points, X and Z, representing the I. C. I. unitary stimuli.

The luminosity coefficients can be assigned any desired values, by use of Eqs. (6). The freedom to assign any shape of the spectrum locus that can be obtained by a projective transformation of the I. C. I. coordinate system is not restricted by the assignment of the luminosity coefficients, except in the case in which $L_u=L_v=L_w$.[11] If the size and shape of the spectrum locus, and the values of the luminosity coefficients are assigned, one further degree of freedom remains in the assignment of the values of the transformation coefficients. When two of the luminosity coefficients are zero, this is a freedom of displacement of the entire coordinate system in a direction parallel to the alychne.

E. Transformation of I. C. I. Data to Modified "Uniform Chromaticity Scale" Diagram

An example of the use of these methods is provided by the following transformation of the

[11] T. Smith, "The Color Triangle and Color Discrimination," Joint Discussion on Vision, Physical and Optical Societies, London, 1932. The special case of equal luminosity coefficients is discussed, beginning on page 217, with the correct statement that the alychne must be projected to infinity. This projection of the alychne to infinity interferes with the freedom to assign the shape of the spectrum locus. The complete discussion of this case would involve some rather annoying exercises in the evaluation of indeterminate ratios, and will be omitted from the present paper.

I. C. I. specifications to another rectangular coordinate system in which the shape of the spectrum locus closely resembles that of Judd's uniform chromaticity scale diagram,[1] while the luminosity coefficients of the corresponding color mixture functions remain $0-1-0$. Since the data on which Judd based his diagram were admittedly approximate in accuracy, some liberties will be taken in modifying the shape of Judd's spectrum locus in order to secure simple transformation equations. If the I. C. I. unitary stimuli are transformed to Judd's coordinate system, it will be found that the ratio ZX/ZY is approximately 10, the angle XZY approximately 90°, the ratio ZP/PX approximately 1/3, and the ratio ZQ/QY is approximately 5. In order to determine the coefficients of the transformation equations, assign:

$u_Z=0$, in order to make $c_3=0$;
$v_Z=0$, $v_X=0$ } in order to make $L_u=L_w=0$, and consequently $c_4=c_6=0$;
$u_Y=0$, in order to make the angle XZY equal to 90°, and consequently $c_2=0$;
$m_X'=1/3$, in order to make $ZP/PX=1/3$, consequently $c_7=-2/3$;
$m_Y'=5$, in order to make $ZQ/QY=5$, consequently $c_8=4$;
$u_X=4.0$, in order to establish the size of the spectrum locus, such that negative tristimulus values are avoided, consequently, $c_1=4/3$;
$v_Y=0.4$, in order to make $ZX/ZY=10$, consequently, $c_5=2.0$.

Substitution of these values of $c_1\cdots c_8$ in Eqs. (3) gives:

$$u=(4/3)x/(-\tfrac{2}{3}x+4y+1),$$
$$v=2y/(-\tfrac{2}{3}x+4y+1). \tag{7}$$

If the numerators and denominator of these equations are multiplied by 3/2, the transformation will assume the simplest possible form:

$$u=2x/(6y-x+1.50),$$
$$v=3y/(6y-x+1.50). \tag{7a}$$

Substitution of the values of $c_1\cdots c_8$ in Eqs. (3a) leads to equations for tristimulus values which are consistent with Eqs. (7) and (7a). The luminosity coefficients of the tristimulus values computed from these equations are $0-2-0$. If all of the coefficients, $a_1\cdots a_9$, are divided by 2, the luminosity coefficients will become $0-1-0$. The condition, $a_3+a_6+a_9=1$, which was introduced solely for convenience in the analysis, is abandoned by this adjustment.

$$U=\tfrac{2}{3}X,$$
$$V=Y,$$
$$W=-0.5X+1.5Y+0.5Z. \tag{8}$$

Since the tristimulus values of the spectrum colors are simply special cases, the color mixture data $\bar{u}_\lambda$, $\bar{v}_\lambda$, $\bar{w}_\lambda$ are derived from the I. C. I. color mixture data $\bar{x}_\lambda$, $\bar{y}_\lambda$, $\bar{z}_\lambda$, using the same transformation coefficients.

Figure 2 shows, as a solid curve, the spectrum locus defined by Eqs. (7a), and shows, as a broken line, the spectrum locus which is secured by a nondistorting transformation of Judd's diagram to rectangular axes and $0-1-0$ luminosity coefficients. The equations which perform the latter transformation are much less

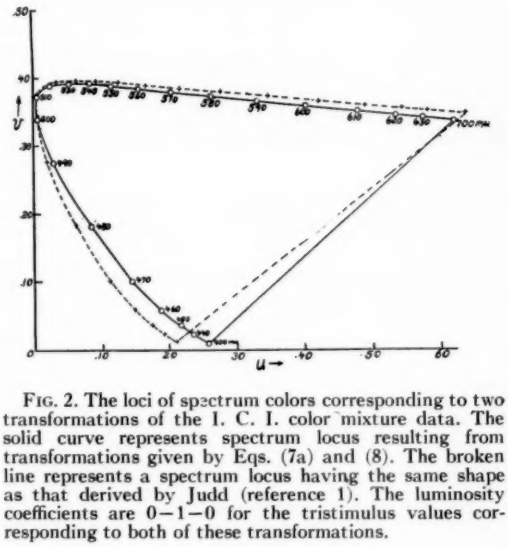

FIG. 2. The loci of spectrum colors corresponding to two transformations of the I. C. I. color mixture data. The solid curve represents spectrum locus resulting from transformations given by Eqs. (7a) and (8). The broken line represents a spectrum locus having the same shape as that derived by Judd (reference 1). The luminosity coefficients are $0-1-0$ for the tristimulus values corresponding to both of these transformations.

convenient than Eqs. (7a), and the distortion to which Eqs. (7a) subject Judd's spectrum locus is negligible in comparison with the probable errors of the data on which Judd's derivation was based.

Figure 3 exhibits the I. C. I. coordinate net transformed according to Eqs. (7a). It is seen that every set of lines which are parallel in the

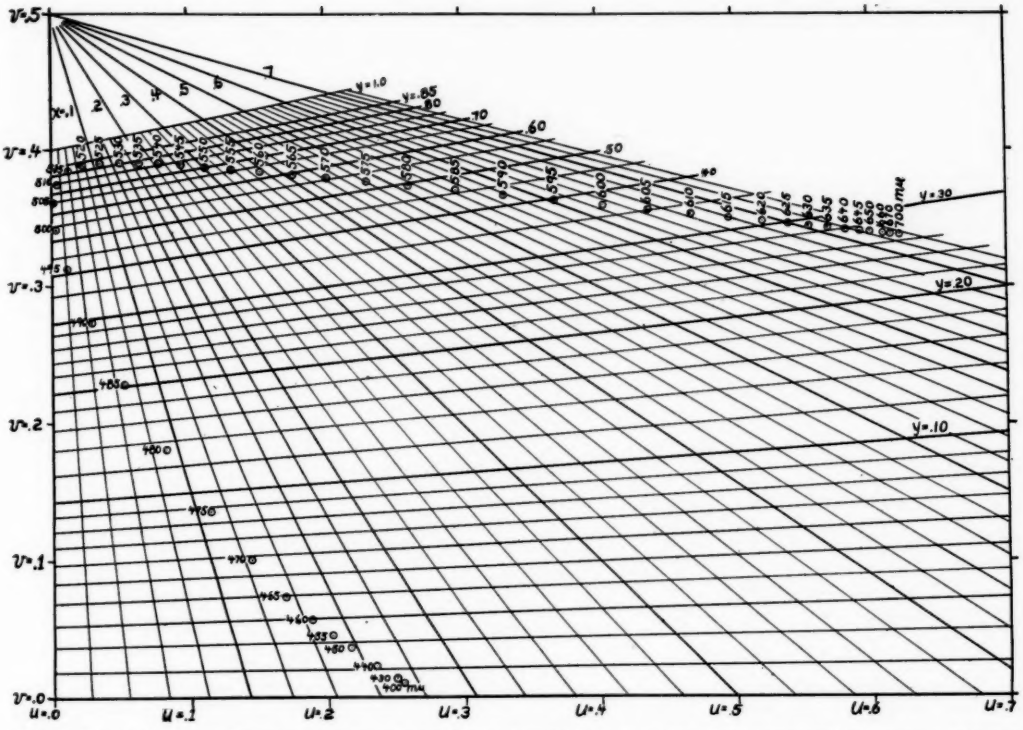

FIG. 3. The I. C. I. coordinate system transformed according to Eqs. (7a). The lines representing constant values of the I. C. I. x coordinate converge through the point $u=0.0$, $v=0.5$. The lines representing constant values of the I. C. I. y coordinate converge through the point $u=-2.0$, $v=0.0$.

I. C. I. diagram is transformed into a set of lines which intersect in a single point in the new coordinate system. The locus of the points of intersection for all sets is a straight line in the new coordinate system. This is one of the general projective properties of such transformations. A familiarity with the basic theorems of projective geometry brings the behavior of transformations of the chromaticity diagram within the scope of intuitive reasoning.

Intensity of Light Reflected from a Parallel Plate

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(Received April 3, 1937)

An equation is derived for the intensity of light reflected from a parallel plate without assuming Stokes' law to hold or a fixed phase change of π on external reflection. It is shown that this equation reduces to equations generally used in describing this intensity only under conditions which are probably never applicable.

INTRODUCTION

THE interference system formed by the reflection of light from a parallel plate is generally described by assuming it to be complementary to the system transmitted through the parallel plate. The equation for the reflected intensity may then be obtained by subtracting from the incident intensity the value derived for the transmitted intensity. The equation usually given[1] for the reflected intensity is of the form

$$I_R=\frac{(4r/(1-r)^2)\sin^2\gamma/2}{1+(4r/(1-r)^2)\sin^2\gamma/2}, \qquad (1)$$

where r is the reflecting power, and γ is the phase difference between successive beams emerging from the plate.

In arriving at the equation for the reflected intensity, Stokes' relations between the reflection and transmission coefficients are generally employed. These relations state in part that the sum

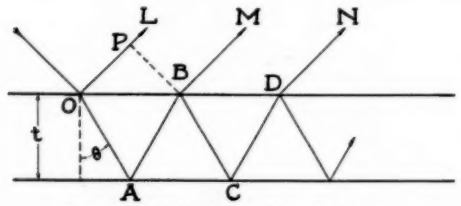

FIG. 1. Multiple beams by reflection.

of the reflectivity and the transmissivity is always unity. They are obviously inapplicable whenever there is any dissipation of energy. At the surface of a glass plate which is metallically coated, there is always an absorption of energy. Many data on this point have been recorded.[2] The sum of the reflective and transmissive powers is always less than 100 percent. It follows that the use of Stokes' relations in describing interference fringes formed by a parallel plate is not justifiable. Furthermore, equations obtained using these relations or their equivalent lead different writers, for example, Planck[3] and Preston[1] to draw opposed conclusions concerning the appearance of these fringes. For these reasons it seems desirable to derive an equation for the reflected intensity from perfectly general considerations.

DERIVATION OF EQUATION FOR REFLECTED INTENSITY

In considering the general case of a parallel plate, having dissimilar surfaces, which produces by multiple reflection two infinite series of beams decreasing in amplitude, let

A_0 = amplitude of the incident beam
a = ratio amplitude is changed at each outside reflection
b = ratio amplitude is changed at each entering refraction
c = ratio amplitude is changed at each inside reflection
d = ratio amplitude is changed at each leaving refraction.

These symbols without subscripts refer to the upper surface, and with subscripts to the lower surface of the plate as shown in Fig. 1. Further, let

$r=cc_1$, the common ratio between successive emergent rays, or the reflecting power

* Now at The Citadel, Charleston, S. C.
[1] See, for example, Preston, *The Theory of Light* (Macmillan and Co., 1928), p. 207.
[2] E.g., v. Murmann, Zeits. f. Physik **80**, 161 (1933); this report gives a comprehensive bibliography.
[3] Planck, *Theory of Light* (Macmillan and Co. 1932), p. 45.

γ = the phase difference between successive emergent rays, given by the expression, $=(2\pi/\lambda)2\mu t\cos\theta$
λ = wave-length of incident light
μ = the refractive index of the plate
t = thickness of the plate
θ = angle of refraction of the incident ray.

The beams transmitted through the plate have respective amplitudes A_0bd_1, $A_0bcc_1d_1$, $A_0bc^2c_1^2d_1$, $\cdots$. If merely for the sake of convenience $A_0=1$, the transmitted series is bd_1, bd_1r, bd_1r^2, $\cdots$. By summing these beams, the complete equation for the transmitted intensity is obtained.[4] This equation is

$$I_t=\frac{b^2d_1^2}{1-2r\cos\gamma+r^2}=\frac{b^2d_1^2}{(1-r)^2}\frac{1}{1+(4r/(1-r)^2)\sin^2\gamma/2}, \quad (2)$$

which is complementary to Eq. (1).

The beams reflected from the plate have respective amplitudes a, bc_1d, bc_1^2cd, $bc_1^3c^2d$, $\cdots$ or a, bc_1d, bc_1dr, bc_1dr^2, $\cdots$. In deriving the intensity function for the reflected system, it is desirable to use the point P in Fig. 1 as a point of reference from which all phase differences must be measured. (BP is perpendicular to OL.) In order to use this point of reference for all beams reflected from the plate, it is necessary to calculate the phase differences which have been introduced into the several beams by traversing different optical paths, or undergoing reflection under different conditions. At the point of incidence, O, all beams which arise by multiple reflection in the plate have the same phase; if P is to be used as the reference point, the origin must be moved from the point where all the beams were alike in phase. Placing the origin at P rather than at O changes the phase of beam OL by an amount $\phi=\alpha+\beta$, where α is the phase difference due to the geometrical path OP, and β is the phase shift arising at external reflection. The transfer of origin changes the phase of BM by an amount $\gamma=\delta-\alpha$, where δ is the advance in phase due to the path OAB, and α is the angle previously defined. Thus γ is the phase due to the difference of optical paths traversed by the successive beams. Beam OL has an amplitude a and a phase β with reference to the origin P; beam BM an amplitude bc_1d and phase γ with respect to the same origin; beam DN, an amplitude bc_1dr and phase 2γ, $\cdots$ and so on for the other beams in the reflected system.

When these beams are superposed by means of a lens, the resulting amplitude produced is the vector sum of all the separate amplitudes superposed. Let σ be the phase with respect to the origin P of the resulting amplitude R. Then

$$R\cos\sigma=a\cos\beta+bc_1d\cos\gamma+bc_1dr\cos 2\gamma+bc_1dr^2\cos 3\gamma+\cdots, \quad (3a)$$

$$R\sin\sigma=a\sin\beta+bc_1d\sin\gamma+bc_1dr\sin 2\gamma+bc_1dr^2\sin 3\gamma+\cdots. \quad (3b)$$

Multiplying the second of these two relations by $i=(-1)^{\frac{1}{2}}$, and adding it to the first,

$$R\epsilon^{i\sigma}=a\epsilon^{i\beta}+bc_1d\epsilon^{i\gamma}+bc_1dr\epsilon^{i2\gamma}+bc_1dr^2\epsilon^{i3\gamma}+\cdots, \quad (4)$$

$$R\epsilon^{i\sigma}=a\epsilon^{i\beta}+\frac{bc_1d\epsilon^{i\gamma}-bc_1dr}{1-r\epsilon^{i\gamma}-r\epsilon^{-i\gamma}+r^2}. \quad (5)$$

By expanding the exponential factors of the right-hand member, simplifying, and then multiplying the resulting equation by its conjugate complex quantity, the equation for the reflected intensity may be obtained. This equation is (since $R^2=I_R$)

$$I_R=a^2+\frac{b^2c_1^2d^2}{1-2r\cos\gamma+r^2}+\frac{2abc_1d[\cos(\beta-\gamma)-r\cos\beta]}{1-2r\cos\gamma+r^2}. \quad (6)$$

Since no assumption is made as to the value of β, Eq. (6) does not demand that a phase shift of π occur at external reflection. β is merely the phase difference introduced by whatever phase shift does occur. This equation, which seems not to be recorded elsewhere, gives a comprehensive description of the reflected intensity.

Discussion

In analyzing Eq. (6) it is justifiable to assume that values of γ which are critical for the trans-

[4] *Handbuch der Physik*, Band XX (J. Springer, Berlin, 1928), p. 13.

mitted system are critical values for the reflected system also. Then, if it is further assumed for an unsilvered plate that β has such a value that $\cos\beta$ is positive, the system formed by reflection is complementary to the system by transmission to the extent that the positions of maxima in one system correspond to the positions of minima in the other. This does not imply that the fringe patterns are complementary. If it is assumed for a silvered plate that β has such a value that $\cos\beta$ is negative, the interference systems are not complementary in any respect. The visibility will be unity for certain values of r in the case of either silvered or unsilvered plates, but, because of the nature of these values, it appears unlikely that zero minima can be observed with a silvered plate.

If, on the other hand, $\beta=0$ be substituted in Eq. (6), the intensity becomes

$$I_R=a^2+\frac{b^2c_1^2d^2+2abc_1d(\cos\gamma-r)}{1-2r\cos\gamma+r^2}, \qquad (7)$$

an equation exactly similar to one which Gilchrist gives[5] without derivation for the case of a silvered plate.

Furthermore, under certain conditions, Eq. (7) can be reduced to Eq. (1). The conditions are that the surfaces of the parallel plate be similar, which makes coefficients $a=a_1$, $b=b_1$, etc., and that Stokes' relations hold, i.e., that $a=-c$ and $bd=1-r$. With these substitutions, Eq. (7) can be simplified to Eq. (1) directly. Unless these conditions prevail it seems impossible to obtain complementary equations—equations such that $I_T+I_R=I$, the incident intensity. The conditions which make possible the reduction from Eq. (6) to Eq. (7) and from Eq. (7) to Eq. (1) are such as to indicate that they are probably never applicable. This fact is probably the strongest argument against the formation of complementary reflected and transmitted fringe systems by a parallel plate, the basis upon which the current explanation depends.

Historical Note

The manner in which these interference systems came to be described as complementary leading to equations such as Eq. (1) and Eq. (2) is a question which naturally arises. About 1833, Airy, making use of previous calculations of Young and Poisson, derived an equation for the intensity of light transmitted through a thin film, presumably in an attempt to explain Newton's rings, which are formed by nonparallel films. Although derived for another purpose, Airy's equation is essentially the same as Eq. (2), which is correct for the parallel plate. It was not until 1849 that reflected and transmitted fringes formed by a parallel plate were first described by Haidinger. In 1906, Lord Rayleigh pointed out[6] that Haidinger's rings rather than Newton's are those directly explained by the usual calculations due to Young, Poisson and Airy. Doubtless, as Lord Rayleigh suggests, the infinity fringes formed by a parallel plate came to be described as complementary by analogy with Newton's rings, which are localized fringes formed by a nonparallel plate.

Acknowledgment

It is an especial pleasure to acknowledge my indebtedness to Mr. Bertrand P. Ramsay of the department of physics, University of Kentucky.

[5] Gilchrist, Phys. Rev. **27**, 596 (1926).

[6] Rayleigh, Phil. Mag. **12**, 490 (1906).

A Comparative Study of Some Violet Absorbing Filters*

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WHEN the blue line of mercury (λ4358) is used for the excitation of Raman spectra, it is often necessary to absorb the violet lines (λ4046 and λ4078). Pfund[1] found that a saturated solution of sodium nitrite strongly absorbs these violet lines. Sannié, Amy, and Poremski[2] studied the absorption of nitrobenzene, sodium acetate, *m*-dinitrobenzene, and many other substances but recommend the first two. Aughey and Lansing[3] found that β-methyl esculetin absorbs the violet portion of the spectrum and may be used in a dry gelatin film. In many cases a liquid filter is objectionable and a stable gelatin filter is desirable. Therefore it seemed advisable to make a study of the spectral absorption of β-methyl esculetin in gelatin films.

β-methyl esculetin is almost insoluble in water, but becomes soluble when one or both of its hydroxyl ions is neutralized. When a solution is first made it is a brilliant lemon yellow. Oxidation is fairly rapid so the esculetin gelatin filter must be dried as quickly as possible. The transmission at λ4358 is materially lowered by oxidation. Gelatin films dyed with β-methyl esculetin were made under many combinations of the following conditions: (1) absorption of the dye in fixed photographic plates, (2) the dyed gelatin solution poured on glass and the dried film stripped off, (3) NaOH, KOH, and NH_4OH used as neutralizing agents, (4) slow and fast drying of films, (5) neutralization of 50 percent (one of the two hydroxyl ions neutralized), 60 percent, and 75 percent, and (6) formalin added to toughen the stripped film. No set of conditions could be found which would consistently give high transmission for the blue line of mercury and a low transmission for the two violet lines. The best films obtained contained one part of the dye to 500 or 1000 parts of gelatin prepared as follows. The dye was 50 percent neutralized with NaOH and dissolved as rapidly as possible, mixed with a warm gelatin solution, and poured onto a glass plate. The gelatin was set quickly by placing it in a cooler and then dried under a strong draft of filtered air. Films made under almost identical conditions showed considerable variation in absorption, due probably to oxidation. Within this wide variation no change in absorption could be found due to the addition of formalin.

In Fig. 1 are shown the transmissions of the best esculetin film which was obtained, of other substances mentioned above, of two samples of Noviol A glass,[4] and of the Wratten 2A gelatin film. It is seen that the Wratten filter surpasses the others in its high transmission at λ4358 compared to that at λ4046 and λ4078, its transmission being 0.1 percent at λ4078 and still less at λ4046. The nitrobenzene solution transmits 0.5 percent at λ4078. The sample used had been fractionally distilled many times at about 2 mm pressure and was much more transparent than that used by Sannié, Amy, and Poremski. However, the absorption at λ4358 varies so much with purity and increases so rapidly under the strong illumination necessary in Raman work that it is not satisfactory. Sannié, Amy, and Poremski recommend a solution of sodium acetate as a satisfactory stable filter. The 40 percent aqueous solution used by the author was perfectly colorless and had but little absorption above λ3500.

Noviol A is satisfactory if its low transmission at λ4358 does not make exposure times too long. As disadvantages, it has the variability common to many glasses, it must be used in flat sheets, and it must be ground quite thin. Saturated sodium nitrite seems to be the best stable liquid filter. However, several centimeters of thickness are required to effectively cut off λ4046. The only convenient way of handling such thick liquid layers seems to be the parallel tube method of R. W. Wood.[5]

* Presented before the Optical Society at New York, Oct. 31, 1936.

[1] Pfund, Phys. Rev. **42**, 581 (1932).

[2] Sannié, Amy, and Poremski, Comptes rendus **202**, 1042 (1936).

[3] Aughey and Lansing, Phys. Rev. **44**, 326 (1933).

[4] Gibson and McNicholas, National Bur. Stand., Tech Papers, No. 119 (1919).

[5] Wood, Phys. Rev. **38**, 2168 (1931).

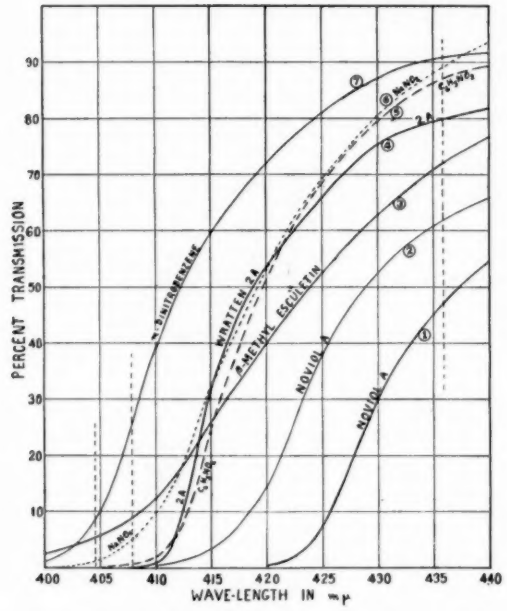

FIG. 1. Spectrophotometric curves. The vertical dotted lines represent the positions of the mercury lines. Curve 1, Noviol A, 2.06 mm thick, National Bureau of Standards data. Curve 2, Noviol A, 2.00 mm thick, National Bureau of Standards data. Curve 3, gelatin film of β-methyl esculetin. Curve 4, Wratten 2A film filter. Curve 5, six percent alcoholic solution of nitrobenzene. Curve 6, saturated water solution of sodium nitrite. Curve 7, two percent alcoholic solution of *m*-dinitrobenzene. Curves 5, 6, and 7 give transmissions of the liquids (the effect of the containing cell being eliminated).

Although the Eastman Kodak Company has found that the Wratten 2A filter is very stable under light from a glass enclosed carbon arc, it seemed desirable to test its stability under mercury arc illumination. Accordingly, several samples were placed inside a glass water-cooling jacket which lay along one focal line of the elliptic reflector of the General Electric Company Raman outfit.[6] A 220 volt hot cathode mercury arc was along the other focal line. The temperature of the samples was kept below 25°C. Transmission curves were made for samples having continuous exposures of 2, 4, 8, and 16 days, approximately 50, 100, 200, and 400 hours. With increasing exposure, the samples showed slightly increased absorption only along the "knee" of the transmission curve from λ4250 to λ4900, the maximum change being near λ4400. No measurable change had occurred during the first 50 hours. At the end of 200 hours there was a 3 percent decrease in the original transmission at λ4358 and at the end of 400 hours a 6 percent decrease. The samples of β-methyl esculetin showed this same type of aging under carbon arc illumination. The amount of decrease of transmission varied greatly from sample to sample, possibly due to moisture content.

[6] Reynolds and Benford, Rev. Sci. Inst. **1**, 413 (1930).

CONCLUSIONS

No gelatin films of β-methyl esculetin were obtained which absorbed well at λ4046. The Wratten 2A film is excellent for absorbing both λ4046 and λ4078. A rapid and very compact Raman apparatus may be made by enlarging one set of holes in a General Electric Raman outfit so as to hold a glass double-walled water cooling tube. The sample tube is wrapped with a layer of the 2A filter and placed inside the cooling tube.

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The author is indebted to Professor C. R. Larkin for the sample of extra-pure nitrobenzene and to Professor V. S. Babasinian for the preparation of the β-methyl esculetin. By controlling the temperature very accurately during the principal reaction and using an additional step of purification, Dr. Babasinian was able to obtain a very pure, almost white, sample. The author is also indebted to Dr. L. A. Jones for allowing the final part of this investigation to be carried on in the Research Laboratories of the Eastman Kodak Company, to E. E. Richardson for all the spectrophotometric measurements, and to E. M. Lowry and Albert Smith for their suggestions and aid in the preparation of the gelatin films of β-methyl esculetin. Discussions with Dr. W. H. Aughey about the properties of the esculetin compound proved very valuable.